

# Effect of Interfacial Structural Phase Transitions on the Coupled Motion of Grain Boundaries: A Molecular Dynamics Study

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## Abstract

In this work the coupled motion of two different phases of  $\Sigma 5(210)[001]$  grain boundaries were investigated by molecular dynamics simulations of fcc Cu. The effect of interfacial structural phase transitions is shown to have a profound effect on both the shear strength and the nature of the coupled motion. Specifically, the motion of the two different phases is described by ideal coupling factors  $\beta^{<100>}$  and  $\beta^{<110>}$  that have different magnitudes and even signs. Additionally, the shear strength for the two interfacial phases is observed to differ by up to 40 % at the lowest temperatures simulated. The study demonstrates that grain boundary phases transitions may have strong effects on the kinetics of microstructural evolution.

Grain boundaries, coupled motion, grain boundary migration, grain boundary structure

Grain boundary (GB) phase transitions are of profound fundamental interest and can have a significant effect on macroscopic properties of materials.[1] Experimental studies suggest that GB phase transitions play important role in abnormal grain growth in ceramics,[2] activated sintering[3] and liquid metal embrittlement.[4] Despite recent progress the effect of these transitions on the kinetics of microstructural evolution is not well understood.

Much of our knowledge about GBs comes from atomistic modeling. Atomistic simulations have provided crucial insights into GB structure and mechanisms of GB migration. Equilibrium and non-equilibrium GB properties like free energies[5, 6, 7, 8] and mobilities[9, 10, 11, 12, 13, 14] were calculated as functions of misorientation and temperature.[15, 16, 17] These properties are often assumed to change continuously with temperature or chemical composition. However, direct studies of the effect of discontinuous changes in GB structure at fixed misorientation on GB migration have not been reported from atomistic simulations to date.

Recent molecular dynamic (MD) simulations reported first order structural transitions in metallic grain boundaries.[18, 19] The discovery of these transitions were enabled by a new simulation methodology. It was demonstrated that the transformations can be triggered by point defects or temperature and gave rise to strong effects on impurity segregation and diffusion.[20] The system studied in Ref. [19] provides a convenient model to investigate the potential role of GB transitions on the kinetics of GB coupled motion.

Coupled GB motion is characterized by a coupling factor  $\beta$ , which is a ratio of the tangential and normal GB velocities. Molecular dynamics studies of the coupled motion of symmetrical tilt  $[100]$  GB in Cu demonstrated two modes of coupling, with ideal coupling factors  $\beta^{<100>}$  and  $\beta^{<110>}$  given by[21]

$$\beta^{<100>} = 2\tan\left(\frac{\theta}{2}\right) \quad (1)$$

$$\beta^{<110>} = -2\tan\left(\frac{\pi}{4} - \frac{\theta}{2}\right) \quad (2)$$

where  $\theta$  is the misorientation angle. It was demonstrated that GBs with  $\theta$  closer to  $0^\circ$  couple in the  $< 100 >$  mode, while GBs with  $\theta$  closer to  $90^\circ$  degrees couple in the  $< 110 >$  mode. The discontinuous transition between the two ideal branches of coupling was observed to occur around  $36^\circ$ . Dual coupling behavior was found for GBs near  $36^\circ$  degrees with the coupling factor spontaneously switching between the two ideal modes during GB motion induced by an imposed constant shear-strain rate in the MD simulations. However, the simulations used periodic boundary conditions with the total number of atoms fixed, thus, prohibiting possible structural transformations of the GBs.

In this work we study coupled motion of different phases of  $\Sigma 5(210)[001]$  GBs in Cu.[22] This is a typical high angle boundary with misorientation angle of  $53.13^\circ$ . The two different atomic structures for this boundary, identified in Ref. [19] and referred to as “split-kite” and “filled-kite” structures, are illustrated in Figure 1 (a) and (b), respectively. The dimensions of the bicrystals with the two GB phases were  $5.1 \times 4.9 \times 16.5 \text{ nm}^3$ , with the  $[001]$  direction parallel  $x$ , and GB planes normal to the  $z$  direction. Due to the difference in GB structures, the number of atoms in the simulation blocks were 33677 and 33576 for split kites and filled kites, respectively. Periodic boundary conditions were applied in the  $x$  and  $y$  directions, while fixed boundary conditions were used in the  $z$  directions to implement shear deformation.[23] Molecular dynamics simulations were performed using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) software package.[24] MD simulations of coupled motion were performed in the temperature range from 0.38-0.98  $T_m$ , where  $T_m=1327 \text{ K}$  is the melting temperature for the Cu potential due to Mishin[22] employed in this work. To investigate the effects of the rate of the deformation, the simulations of coupled motion were performed with shear rates spanning an order of magnitude, with imposed shear velocities of 1m/s and 0.1 m/s.

Figure 2 (a) illustrates the bicrystals with split-kites and filled-kite GB phases after 30 ns of the coupled motion. The initial positions of the GBs are indicated by the white dashed lines. To track the shape changes of the bicrystals, a column of atoms that were initially vertical are colored in red. It is evident from the figure that the part of the block swept by the GBs is sheared. While the upper grains of both bicrystals moved to the right by the same amount as indicated by the white arrows, the split kites GB (left) moved up and filled kites (right) moved down. The amounts of the normal displacement observed for the same simulation time are also different.

Figures 2 (b) and (c) show typical plots of the shear stress and the GB displacements as functions of time at 700 K for split kites and filled kites, respectively. Both phases move by characteristic stick-slip dynamics, however the direction and magnitude of GB displacements as well as the shear stresses are different for the two GB phases.

From the knowledge of the imposed shear rate and the observed normal displacements (Figures 2 (b) and (c)) the value of the GB coupling factor  $\beta = v_{\parallel}/v_{\perp}$  was calculated from MD simulations. Figure 3 shows the inverse coupling factors  $1/\beta$  for the two GB phases as functions of temperature. The blue and red horizontal lines on the plot represent values of the ideal coupling factors  $\beta^{<100>}$  and  $\beta^{<110>}$ , calculated using Eqs. (1) and (2). It is evident from the figure that until about 0.83  $T_m$  the split kite and filled kite phases move with ideal coupling factors, corresponding to the  $< 100 >$  and  $< 110 >$  branches, respectively. For this range of temperature and shear

rates the GB structures are preserved during the coupled motion and no spontaneous switching between modes of coupling was observed. The absence of switching is different from results reported in previous studies[23] indicating that the coupling behavior is sensitive to the details of the atomistic GB structure.

At temperatures above  $0.83 T_m$  coupling factors of both GBs become non-ideal. The filled-kite phase shows increasing sliding until it completely premelts at 1300 K, and the motion of the GB is purely sliding in nature. The split-kite phase also becomes increasingly disordered with its structural units disappearing around 1200 K. This change in the GB structure correlates with the observation of a non-ideal value and change in sign for the coupling factor  $\beta$ .

Finally, the average shear stress during coupled motion was calculated for the two GB phases and is plotted as a function of temperature in Figure 4. While both stresses decrease with temperature, the stress required to move filled kites is only about 60% of that for split kites. This shows that given the same driving force GB transitions can noticeably increase or slow down GB motion.

The GB phases studied here differ significantly in atomic density, and as a result the kinetics of these transitions are limited by diffusion of atoms. It is thus quite possible that different GB phases may be present simultaneously in equilibrium with each other or due to slow kinetics. The mechanism of motion of such boundaries is currently not clear, since different segments of the boundary will move in opposite directions in the response to the same driving force. As a result, such mixed-phase GBs can become less mobile with one of the phases dragging the other.

This study also motivates a systematic investigation of coupled motion of different phases of other high angle GBs, to see whether the simultaneous presence on different branches of coupling depending on GB structure is a common phenomenon. Experimental measurements of coupling factor for Al showed an abrupt transition from  $\langle 100 \rangle$  to  $\langle 110 \rangle$  modes with increasing misorientation,[26] confirming results the previous MD studies for Cu.[23] The current investigation shows that the switching of the GB coupling mode for the same grain misorientation can be triggered by GB phase transitions. Thus, the study suggests a possible indirect way to discover GB phase transitions by measuring variations in the coupling factor at different temperatures and impurity levels.

In summary, coupled motion of split-kite and filled-kite phases of a  $\Sigma 5(210)$  GB in Cu was investigated using atomistic simulations. Both the coupling mode and GB shear strength were observed to depend on GB phase for fixed misorientation angle. Specifically, the motion of the phases is described by ideal coupling factors  $\beta^{\langle 100 \rangle}$  and  $\beta^{\langle 110 \rangle}$  that have different magnitudes and even signs. Thus, the present simulation results demonstrate clearly that GB structural phase transitions have a significant effect on kinetic properties of GBs and in some cases may even reverse the direction of GB motion.[25]

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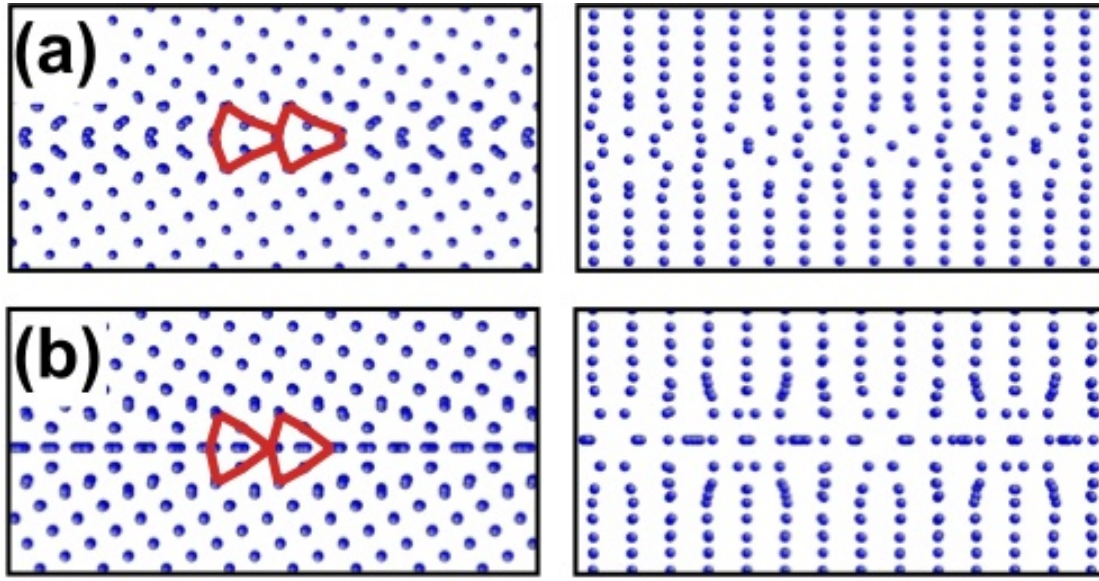


Figure 1: (a) Split-kite and (b) filled-kite phases of  $\Sigma 5(210)$  grain boundary calculated at 0K in Ref. [19] using an EAM potential for Cu.[22]

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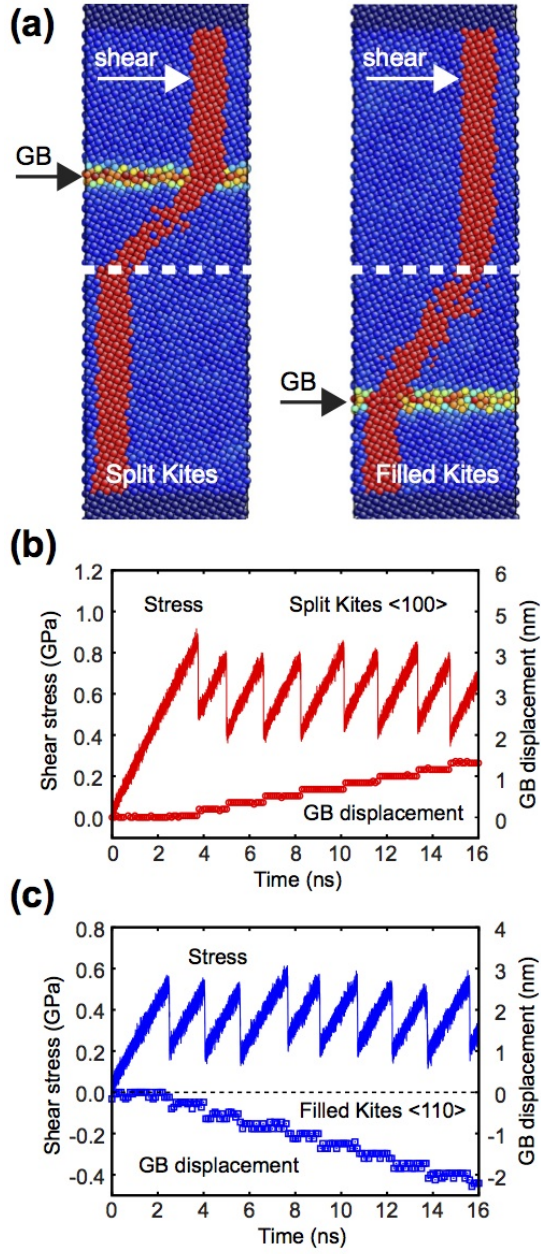


Figure 2: (a) Bicrystals with split kites (left) and filled kites (right) GB phases after 30 ns of coupled motion at 700 K with  $v_{\parallel} = 0.1$  m/s. Initially vertical, red stripe serves to illustrate the shear deformation. Shear stress and grain boundary displacement during coupled motion of split-kite (b) and filled-kite (c) phases.

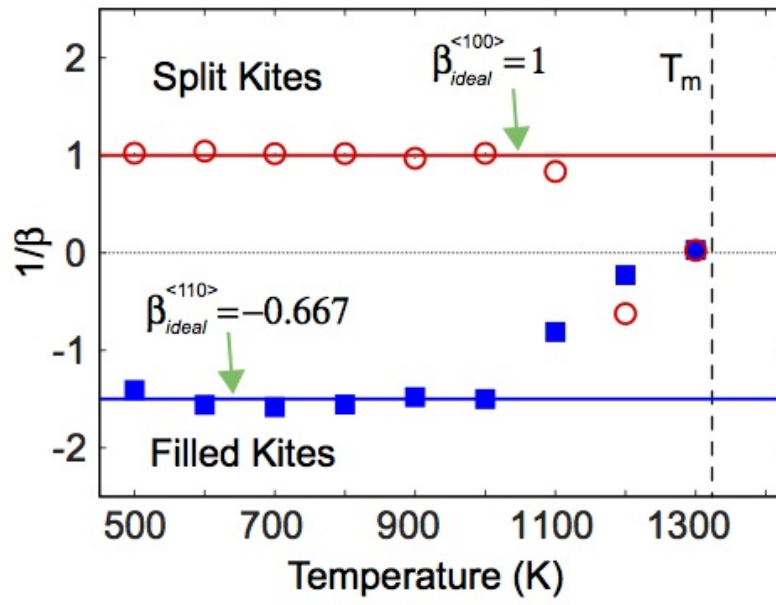


Figure 3: Inverse coupling factor  $1/\beta$  as function of temperature for split kites (red circles) and filled kites (blue squares). Solid horizontal lines correspond to ideal values  $1/\beta^{<100>}$  and  $1/\beta^{<110>}$  calculated using Eqs. (1) and (2). The vertical dashed line indicates the bulk melting temperature  $T_m = 1327$  K.

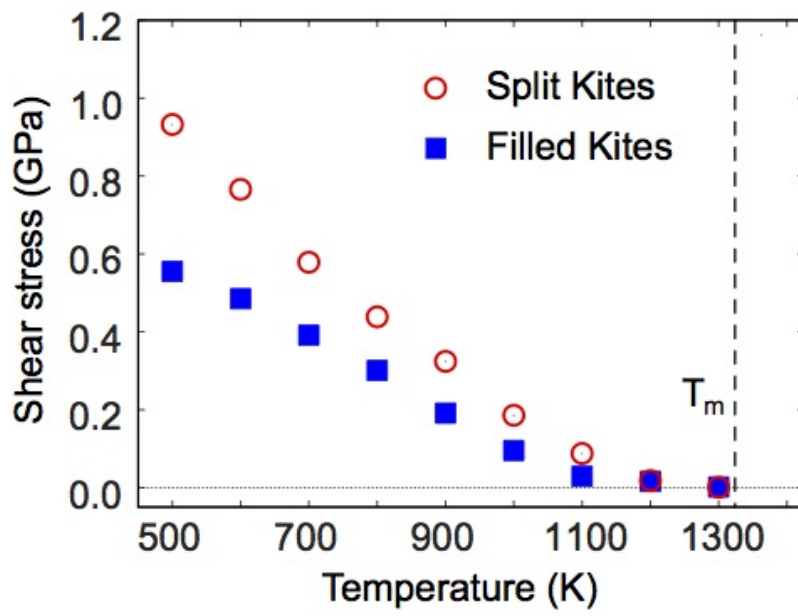


Figure 4: Shear stress as a function of temperature for split kites (red circles) and filled kites (blue squares) for  $v_{\parallel} = 1$  m/s. The vertical dashed line indicates the bulk melting temperature  $T_m = 1327$  K.